View metadata, citation and similar papers at core.ac.uk

brought to you by CORE

provided by Elsevier - Publisher Connector

Arabian Journal of Chemistry (2016) 9, S663–S667

King Saud University

Arabian Journal of Chemistry

www.ksu.edu.sa
www.sciencedirect.com

ORIGINAL ARTICLE

# PTA ($H_3PO_4 \cdot 12WO_3 \cdot xH_2O$): An eco-friendly catalyst for the synthesis of new Schiff-bases containing benzimidazole moiety

**Varsha U. Vernekar, Kallappa M. Hosamani *, Irfan N. Shaikh, Kavitha C.S. Achar**

*P.G. Department of Studies in Chemistry, Karnatak University, Pavate Nagar, Dharwad 580 003, India*

Received 27 May 2011; accepted 22 July 2011
Available online 30 July 2011

**KEYWORDS**

Benzimidazole derivatives;
3-Amino-2-naphthoic acid;
Schiff's base derivatives;
Aldehydes;
$H_3PO_4 \cdot 12WO_3 \cdot xH_2O$

**Abstract** In this study $H_3PO_4 \cdot 12WO_3 \cdot xH_2O$ is found to catalyze the preparation of Schiff bases from the reaction of 3-(1*H*-Benzimidazol-2-yl)naphthalene-2-amine with different aldehydes efficiently in ethanol. The advantages of this environmental friendly and mild method are such as simplicity of the reaction procedure, the elimination of solvents, simple work-up, high product yields and short reaction times. The products were characterized by FT-IR $^1$H NMR, $^{13}$C NMR, mass spectrometry and elemental analysis.

© 2011 Production and hosting by Elsevier B.V. on behalf of King Saud University. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/3.0/).

## 1. Introduction

In recent years, the use of heterogeneous catalysts have received considerable interest in various disciplines, including organic synthesis owing to their easy work-up procedures, easy filtration, commercially available, environmentally friendly, minimization of cost and no effluent generation due to recycling of these catalysts (Gadekara et al., 2009; Clark, 2002; Alizadeh et al., 2003). The applications of heterogeneous catalyst, especially heteropolyacids have become most important for the development of environmentally benign chemical processes and technologies in green chemistry (Heravi et al., 2008; Clark, 2001; Anastas and Warner, 1998). Heteropolyacids have been found to act as outstanding catalysts in electrophillic transformation. They are stronger than the usual mineral acids, such as HCl, $H_2SO_4$ and $HNO_3$ (Heravi and Sadjadi, 2009). Among the heteropolyacids, PTA is the most widely used catalyst, because of its high acid strength, thermal stabilities and low reducibilities (Devassy et al., 2004).

Benzimidazole ring plays an important heterocyclic pharmacophore in drug discovery. These compounds carrying out different substituents in benzimidazole structure are associated with a wide range of biological activities including anticancer (Kruse et al., 1989), antiviral (Townsend, 1996), antimicrobial (Hubschwerlen et al., 1992), antihelmentic (Veerakumari and Munuswamy, 2000), anti-inflammatory (Labanauskas et al., 2000), antihistaminic (Iemura et al., 1986), proton pump

* Corresponding author. Tel.: +91 836 2215286; fax: +91 836 2771275, +91 836 2747884.
E-mail address: dr_hosamani@yahoo.com (K.M. Hosamani).

Peer review under responsibility of King Saud University.

Production and hosting by Elsevier

http://dx.doi.org/10.1016/j.arabjc.2011.07.030

1878-5352 © 2011 Production and hosting by Elsevier B.V. on behalf of King Saud University.
This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/3.0/).

inhibitor (Kuhler et al., 1988), antioxidant (Cole et al., 1974), antihypertensive (Kubo et al., 1993) and anticoagulant (Mederski et al., 2004) properties. These derivatives also exhibit significant activity against several viruses such as HIV (Porcari et al., 1998; Roth et al., 1997).

Schiff bases derived from aromatic amines and aromatic aldehydes have a wide variety of applications in many fields, e.g., biological, inorganic and analytical chemistry (Cimerman et al., 2000; Singh et al., 1975; Perry et al., 1988). They are reported to show characteristic biological activities including antibacterial, antifungal, anticancer, anti-convulsant anti-inflammatory activities (Jarrahpour et al., 2006; Taggi et al., 2002a,b; Chohan et al., 2006; Pandeya et al., 1999; Pathak et al., 2000; Samadhiya and Halve, 2001). Other applications of Schiff's bases include industrial synthesis of high value life saving beta lactam Taggi et al., 2002a,b antibiotics from a class of penicillins and cephalosporins. Owing to the importance of PTA and in continuation of our work on the development of eco-friendly reactions, we report synthesis of new Schiff bases containing benzimidazole moiety with various aromatic aldehydes by using PTA as an eco-friendly catalyst.

## 2. Results and discussions

In the present protocol, we are reporting a simple method for the synthesis of Schiff's base from substituted 3-(1*H*-Benzimidazol-2-yl) naphthalene-2-amine. Initially substituted 3-(1*H*-Benzimidazol-2-yl) naphthalene-2-amine is synthesized by the reaction of 3-amino-2-naphthoic acid and substituted *o*-phenylenediamine in the presence of PTA as catalyst to afford substituted 3-(1*H*-Benzimidazol-2-yl) naphthalene-2-amine. Later the free $NH_2$ was condensed with various aldehydes to give Schiff bases. The purity of the synthesized compounds was checked by performing TLC. Using optimized reaction parameters, Schiff base derivatives **(a11–a20)** were synthesized. The time required and the yield of the products is given in Table 1. In each case, condensation of substrates occurred and upon work-up, the corresponding Schiff base was obtained. As seen in Table 1, a lot of Schiff bases were synthesized using PTA as a catalyst in ethanol.

The reaction proceeds rapidly with 5 mol% of catalyst and is complete within 2 h. In a control experiment, it was observed that in the absence of the catalyst, reaction did not proceed even at higher temperatures. It is of great importance that the reaction is largely affected by the PTA catalyst. The entries 3 and 9, (Table 1) respectively, show the reaction of *p*-nitro-benzaldehyde with 3-(1*H*-benzo[d]imidazole-2-yl)naphthalen-2-amine and the reaction of *p*-nitro-benzaldehyde with 3-(6-methyl-1*H*-benzo[d]imidazole-2-yl)naphthalen-2-amine under reflux in ethanol solution with long reaction times, the corresponding products were obtained in low yields. This low yield may be related to steric hindrance in benzaldehyde accompanied to electron withdrawing of nitro group on aromatic ring of aldehyde.

**Table 1** The reaction of amine with various aldehydes catalyzed by $H_3PO_4.12WO_3.xH_2O$ in Ethanol.

| Entry | $R^1$ | Time (h) | Compounds | Yield (%) | m.p. (°C) |
|---|---|---|---|---|---|
| 1 | H | 3 | a11 | 83 | 142–144 |
| 2 | 2-Cl | 2 | a12 | 79 | 196–197 |
| 3 | 2-$NO_2$ | 3 | a13 | 42 | 140–141 |
| 4 | 3-Cl | 2.5 | a14 | 79 | 178–180 |
| 5 | 4-$OCH_3$ | 1.5 | a15 | 69 | 156–158 |
| 6 | H | 2 | a16 | 83 | 121–122 |
| 7 | 4-$OCH_3$ | 2 | a17 | 66 | 104–106 |
| 8 | 2-Cl | 1.5 | a18 | 80 | 75–76 |
| 9 | 2-$NO_2$ | 3 | a19 | 46 | 106–108 |
| 10 | 3-Cl | 3 | a20 | 82 | 133–134 |

**Table 2** Solvent effect on the synthesis of Schiff bases.[a]

| Entry | Solvents | Time (h)[b] | Yield (%)[c] |
|---|---|---|---|
| 1 | DMF | 12 | 43 |
| 2 | 1,4-Dioxane | 14 | 58 |
| 3 | THF | 8 | 38 |
| 4 | Methanol | 7 | 51 |
| 5 | DMSO | 26 | 60 |
| 6 | Ethanol | 1.5 | 95 |

[a] The mixture of 3-(1*H*-Benzimidazol-2-yl) naphthalene-2-amine and benzaldehyde using PTA as catalyst in different solvents.
[b] Time to finish the reaction monitored by TLC.
[c] Yield refers to isolated products.

In order to evaluate the effect of the solvent, reaction was carried out with 3-(1*H*-Benzimidazol-2-yl) naphthalene-2-amine and benzaldehyde using PTA as a model reaction (Table 2). When the reaction was performed in ethanol using PTA as catalyst, the reaction was very fast and the product was easily isolated from the reaction mixture. The model reaction was also performed using different solvents like DMF, 1,4-dioxane, THF, methanol, DMSO (Table 2). But the yields were low and the reaction times were too long. These results suggest that ethanol is the best solvent for this method.

The structures of the synthesized compounds were determined on the basis of their FT-IR and $^1$H NMR data and elemental analysis. The IR spectra of the synthesized compounds showed the presence of C═N stretching bands around 1687–1612 $cm^{-1}$ and NH stretching frequencies appear in the range of 3435–3324 $cm^{-1}$ corresponding to azomethine compounds. In the $^1$H NMR spectra of the target schiff's base, the –NH proton of imidazole ranges from 9.71–11.04 ppm. While N═CH proton varies from 4.38–4.57 ppm. The other protons appeared at the expected chemical shifts.

## 3. Conclusions

In conclusion, a successful attempt has been made to synthesize Schiff's base derivatives from corresponding substituted 3-(1*H*-Benzimidazol-2-yl) naphthalene-2-amine and various aldehydes using ethanol as solvent in high yields. The method employed is very simple and economically viable.

## 4. Experiments

### 4.1. Methods and materials

All compounds were routinely checked by thin-layer chromatography (TLC) on aluminum-backed silica gel plates. Melting points were determined with open capillary method and are uncorrected. IR spectra were recorded on a *Nicolet 5700 FT-IR* instrument (*Nicolet, Madison, WI, USA*) as KBr discs. The $^1$H-NMR and $^{13}$C-NMR spectra were measured with a

*Bruker-300* (*Bruker Bioscience, USA*), 300 MHz instrument using DMSO as solvent and TMS as internal standard. All chemical shifts were reported as $\delta$ values (ppm). Mass spectrometer with ionization energy maintained at 70ev using *Shimadzu Mass Spectrometer (LCMS)* and elemental analysis were carried out on *MT-3 analyzer*.

### 4.2. General procedure and spectral data of stage I

A mixture of *o*-phenylenediamine (0.01 mol) and 3-amino-napthoic acid (0.01 mol) was taken in RB flask, add 20 ml of 6 N HCl and then refluxed for 6 h at about 120 °C. After completion of the reaction monitored by TLC (Eluent: 2:1 *n*-Hexane/Ethyl acetate) the reaction mixture is poured in crushed ice and basified with ammonia, filtered and recrystallised from methanol and was washed with water. Then the solid product was filtered off and recrystallized from DMF.

#### 4.2.1. 3-(1H-benzo[d]imidazole-2-yl)naphthalen-2-amine (**a**)

Yield 76%; mp 179–180 °C; IR (KBr) ($\nu_{max}/cm^{-1}$): 3372 (NH of imidazole), 2913 (=C–H); $^1$H NMR (300 MHz, DMSO-$d_6$) $\delta$ 4.55 (s, 2H, $NH_2$) 6.99 (s, 1H, Ar–H), 7.09–7.34 (s, 4H, Ar–H), 7.50–7.77 (m, 5H, Ar–H), 10.14 (s, 1H, NH of imidazole); $^{13}$C NMR (75 MHz, DMSO) $\delta$ 107.6 (Ar–C), 117.3 (Ar–C), 124.1 (Ar–C), 152.6 (C of imidazole). LC–MS: *m*/*z* 260.36 ($M^+$). Anal. calcd for $C_{21}H_{15}N_5O_3$: C, 78.74; H, 5.05; N, 16.20%. Found: C, 70.71; H, 5.03; N, 16.23%.

#### 4.2.2. 3-(6-methyl-1H-benzo[d]imidazole-2-yl)naphthalen-2-amine (**b**)

Yield 64%; mp 153–155 °C; IR (KBr) ($\nu_{max}/cm^{-1}$): 3432 (NH of imidazole), 2923 (=C–H), $^1$H NMR (300 MHz, DMSO-$d_6$) $\delta$ 2.50 (s, 3H, $CH_3$) 4.55 (s, 2H, $NH_2$) 7.00 (d, 1H, Ar–H), 7.11 (s, 1H, Ar–H), 7.37–7.39 (d, 2H, Ar–H), 7.50 (s, 2H, Ar–H), 7.53–7.75 (d, 3H, Ar–H), 9.98 (s, 1H, NH of imidazole); $^{13}$C NMR (75 MHz, DMSO) $\delta$ 110.3 (Ar–C), 114.6 (Ar–C), 127.1 (Ar–C), 150.6 (C of imidazole). LC–MS: *m*/*z* 272.48 (M). Anal. calcd for $C_{21}H_{15}N_5O_3$: C, 79.10; H, 5.53; N, 15.37%. Found: C, 79.13; H, 5.50; N, 15.35%.

### 4.3. General procedure and spectral data of stage II

A mixture of substituted 3-(1*H*-Benzimidazol-2-yl) naphthalene-2-amine (0.005 mol), various aldehydes (0.005 mol) and PTA (5 mol%) in absolute alcohol was heated under reflux for 16 h on water bath. After the completion of reaction as indicated by TLC (Eluent: 1:1 Cyclohexane/Ethyl Acetate) the reaction mixture was poured in crushed ice and the solid was filtered off. The product obtained was recrystallized from 1:1 DMF and Ethanol.

#### 4.3.1. 3-(1H-benzo[d]imidazol-2-yl)-N-benzylidenenaphthalen-2-amine (**a11**)

Light Green Solid, IR (KBr) ($\nu_{max}/cm^{-1}$): 3373 (NH of imidazole), 2922 (=C–H), 1687(C=N); $^1$H NMR (300 MHz, DMSO-$d_6$) $\delta$ 4.56 (s, 1H, N=CH), 6.93–7.18 (d, 4H, Ar–H), 7.36–7.61 (s, 5H, Ar–H), 7.68–7.92 (s, 6H, Ar–H), 9.98 (s, 1H, NH of imidazole); $^{13}$C NMR (75 MHz, DMSO) $\delta$ 116.5 (Ar–C), 124.3 (Ar–C), 128.9 (Ar–C), 150.6 (C of imidazole), 161.8 (HC=N). LC–MS: *m*/*z* 346.23 (M). Anal. calcd for $C_{21}H_{15}N_5O_3$: C, 82.97; H, 4.93; N, 12.10%. Found: C, 83.00; H, 3.93; N, 18.17%.

#### 4.3.2. 3-(1H-benzimidazol-2-yl)-N-(2-chlorobenzylidine)naphthalen-2-amine (**a12**)

Green Colored Solid, IR (KBr) ($\nu_{max}/cm^{-1}$): 3373(NH of imidazole), 2924 (=C–H), 1662 (C=N); $^1$H NMR (300 MHz, DMSO-$d_6$) $\delta$ 4.39 (s, 1H, N=CH), 6.99–7.11 (d, 4H, Ar–H), 7.36–7.52(s, 4H, Ar–H), 7.74–8.14(s, 6H, Ar–H), 10.25(s, 1H, NH of imidazole); $^{13}$C NMR (75 MHz, DMSO) $\delta$ 113.3 (Ar–C), 123.6 (Ar–C), 128.4 (Ar–C), 134.7 (C–Cl), 150.8 (C of imidazole) 163.4 (HC=N). LC-MS: *m*/*z* 381 (M), Anal. calcd for $C_{21}H_{15}N_5O_3$: C, 75.49; H, 4.22; N, 11.00%. Found: C, 75.46; H, 4.24; N, 11.03%.

#### 4.3.3. 3-(1H-benzimidazol-2-yl)-N-(2-nitrobenzylidene) naphthalen-2-amine (**a13**)

Dark Yellow Color, IR (KBr) ($\nu_{max}/cm^{-1}$): 3375 (NH of imidazole), 2853 (=C–H), 1627 (C=N), 1339 and 1570 ($NO_2$ symm and asymm); $^1$H NMR (300 MHz, DMSO-$d_6$) $\delta$ 4.46 (s, 1H, N=CH), 7.00–7.11 (d, 4H, Ar–H), 7.50–7.75 (s, 4H, Ar–H), 7.77–7.95 (s, 6H, Ar–H), 10.13 (s, 1H, NH of imidazole); $^{13}$C NMR (75 MHz, DMSO) $\delta$ 117.8 (Ar–C), 122.5 (Ar–C), 127.4 (Ar–C), 147.7 (C–$NO_2$), 151.1 (C of imidazole) 162.3 (HC=N). LC–MS: *m*/*z* 392.46 (M), Anal. calcd. for $C_{21}H_{15}N_5O_3$: C, 73.46; H, 4.11; N, 14.28%. Found: C, 73.48; H, 4.13; N, 14.25%.

#### 4.3.4. 3-(1H-benzimidazol-2-yl)-N-(3-chlorobenzylidine) naphthalen-2-amine (**a14**)

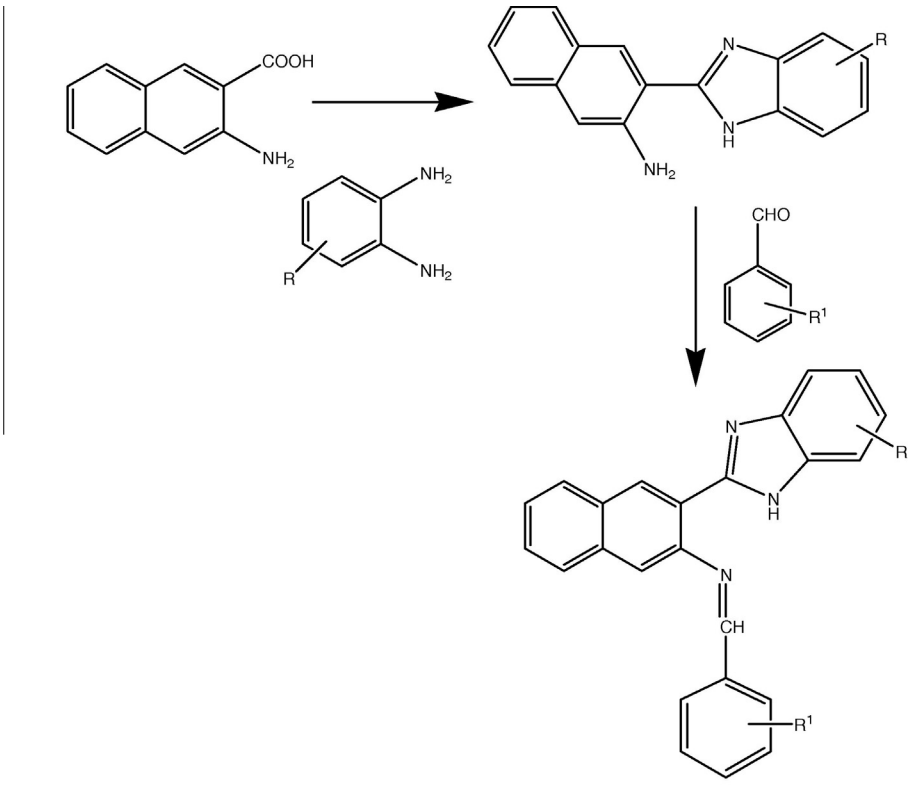
Dark Green Solid, IR (KBr) ($\nu_{max}/cm^{-1}$): 3374 (NH of imidazole), 2849(=C–H), 1629 (C=N); $^1$H NMR (300 MHz, DMSO-$d_6$) $\delta$ 4.53 (s, 1H, N=CH), 6.99–7.11 (d, 4H, Ar–H), 7.37–7.52(s, 4H, Ar–H), 7.75–7.95 (s, 6H, Ar–H), 9.79 (s, 1H, NH of imidazole); $^{13}$C NMR (75 MHz, DMSO) $\delta$ 115.3 (Ar–C), 124.2 (Ar–C), 129.9 (Ar–C), 133.3 (C–Cl), 152.5 (C of imidazole) 160.8 (HC=N). LC–MS: *m*/*z* 379.99 (M), Anal. calcd for $C_{21}H_{15}N_5O_3$: C, 75.49; H, 4.22; N, 11.00%. Found: C, 75.47; H, 4.25; N, 10.58%.

#### 4.3.5. 3-(1H-benzimidazol-2-yl)-N-(4-methoxybenzylidine) naphthalen-2-amine (**a15**)

Green Colored Solid, IR (KBr)($\nu_{max}/cm^{-1}$): 3324 (NH of imidazole), 2923(=C–H), 1629(C=N); $^1$H NMR (300 MHz, DMSO-$d_6$) $\delta$ 4.49 (s, 1H, N=CH), 3.87 (s, 3H, O–$CH_3$), 7.00–7.12 (d, 4H, Ar–H), 7.38–7.52(s, 4H, Ar–H), 7.78–7.89 (s, 6H, Ar–H), 10.36 (s, 1H, NH of imidazole); $^{13}$C NMR (75 MHz, DMSO) $\delta$ 58.3 (O–$CH_3$), 112.6 (Ar–C), 122.5 (Ar–C), 129.3 (Ar–C), 155.7 (C of imidazole) 159.8 (HC=N). LC–MS: *m*/*z* 378.33 ($M^+$), Anal. calcd for $C_{21}H_{15}N_5O_3$: C, 79.55; H, 5.07; N, 11.13%. Found: C, 79.58; H, 5.10; N, 11.11%.

#### 4.3.6. 3-(5-methyl-1H-benzimidazol-2-yl)-N-(benzylidene) naphthalen-2-amine (**a16**)

Brown Colored Solid, IR (KBr)($\nu_{max}/cm^{-1}$): 3435 (NH of imidazole), 2850(=C–H); 1632 (C=N); $^1$H NMR (300 MHz, DMSO-$d_6$) $\delta$ 2.43 (s, 3H, Ar–$CH_3$), 4.57 (s, 1H, N=CH), 7.00–7.21 (d, 3H, Ar–H), 7.37–7.51 (s, 5H, Ar–H), 7.62–8.14 (s, 6H, Ar–H), 9.72 (s, 1H, NH of imidazole); $^{13}$C NMR (75 MHz, DMSO) $\delta$ 26.3 (Ar–$CH_3$), 114.9 (Ar–C), 122.8 (Ar–C), 127.6 (Ar–C), 150.1 (C of imidazole) 162.5 (HC=N). LC–MS: *m*/*z* 360.36 (M), Anal. calcd. for $C_{21}H_{15}N_5O_3$: C, 83.08; H, 5.30; N, 11.63%. Found: C, 83.10; H, 5.27; N, 11.66%.




R= H, $CH_3$
$R^1$= H, 2-Cl, 3-Cl, 2-$NO_2$, 4-$OCH_3$

**Scheme 1** Schematic representation of 3-(1*H*-benzo[d] imidazol-2-yl)-*N*- benzylidenenaphthalen-2-amine derivatives.

*4.3.7. 3-(5-methyl-1H-benzimidazol-2-yl)-N-(4-methoxybenzylidene)naphthalen-2-amine* ***(a17)***
Yellow Colored Solid, IR (KBr)($\nu_{max}$/$cm^{-1}$): 3351 (NH of imidazole), 2918(=C–H), 1631 (C=N); $^1$H NMR (300 MHz, DMSO-$d_6$) $\delta$ 2.45 (d, 3H, Ar–$CH_3$), 3.73 (s, 3H, O–$CH_3$), 4.41 (s, 1H, N=CH), 6.91–7.14 (d, 3H, Ar–H), 7.41–7.50 (s, 4H, Ar–H), 7.78–8.11 (s, 6H, Ar–H), 11.04 (s, 1H, NH of imidazole); $^{13}$C NMR (75 MHz, DMSO) $\delta$ 24.8 (Ar–$CH_3$), 56.2 (O–$CH_3$), 116.8 (Ar–C), 124.9 (Ar–C), 128.3 (Ar–C), 150.8 (C of imidazole) 160.2 (HC=N). LC–MS: *m*/*z* 392.21 ($M^+$), Anal. calcd. for $C_{21}H_{15}N_5O_3$: C, 79.77; H, 5.41; N, 10.73%. Found: C, 79.80; H, 5.43; N, 10.70%.

*4.3.8. 3-(5-methyl-1H-benzimidazol-2-yl)-N-(2-chlorobenzylidene)naphthalen-2-amine* ***(a18)***
Yellow Colored Solid, IR (KBr)($\nu_{max}$/$cm^{-1}$): 3354 (NH of imidazole), 2852 (=C–H), 1631 (C=N); $^1$H NMR (300 MHz, DMSO-$d_6$) $\delta$ 2.50 (d, 3H, Ar–$CH_3$), 4.52 (s, 1H, N=CH), 6.83–7.15 (d, 3H, Ar–H), 7.42–7.60 (s, 4H, Ar–H), 7.78–8.15 (s, 6H, Ar–H), 10.36 (s, 1H, NH of imidazole); $^{13}$C NMR (75 MHz, DMSO) $\delta$ 26.1 (Ar–$CH_3$), 115.8 (Ar–C), 125.2 (Ar–C), 127.6 (Ar–C), 134.2 (C–Cl), 154.5 (C of imidazole) 162.9 (HC=N). LC–MS: *m*/*z* 396.02 (M), Anal. calcd. for $C_{21}H_{15}N_5O_3$: C, 75.85; H, 4.58; N, 10.61%. Found: C, 75.82; H, 4.56; N, 10.64%.

*4.3.9. 3-(5-methyl-1H-benzimidazol-2-yl)-N-(2-nitrobenzylidene)naphthalen-2-amine****(19)***
Dark Brown Solid, IR (KBr)($\nu_{max}$/$cm^{-1}$): 3426 (NH of imidazole), 2918 (=C–H),1612 (C=N), 1357 and 1528 ($NO_2$ symm and asymm); $^1$H NMR (300 MHz, DMSO-$d_6$) $\delta$ 2.51 (d, 3H, Ar–$CH_3$), 4.38 (s, 1H, N=CH), 7.06–7.12 (d, 3H, Ar–H), 7.39–7.75 (s, 4H, Ar–H), 7.86–8.22 (s, 6H, Ar–H), 9.71 (s, 1H, NH of imidazole); $^{13}$C NMR (75 MHz, DMSO) $\delta$ 22.3 (Ar–$CH_3$), 113.8 (Ar–C), 124.0 (Ar–C), 127.6 (Ar–C), 149.2 (C–$NO_2$), 152.7 (C of imidazole) 164.6 (HC=N). LC–MS: *m*/*z* 407.14 (M), Anal. calcd. for $C_{21}H_{15}N_5O_3$: C, 73.88; H, 4.46; N, 13.78%. Found: C, 73.91; H, 4.48; N, 13.75%.

*4.3.10. 3-(5-methyl-1H-benzimidazol-2-yl)-N-(3-chlorobenzylidene)naphthalen-2-amine* ***(a20)***
Black Colored Solid, IR (KBr)($\nu_{max}$/$cm^{-1}$): 3434 (NH of imidazole), 2921 (=C–H), 1631 (C=N); $^1$H NMR (300 MHz, DMSO-$d_6$) $\delta$ 2.09 (d, 3H, Ar–$CH_3$), 4.54 (s, 1H, N=CH), 6.85–7.06 (d, 3H, Ar–H), 7.38–7.55 (s, 4H, Ar–H), 7.78–8.10 (s, 6H, Ar–H), 10.18 (s, 1H, NH of imidazole); $^{13}$C NMR (75 MHz, DMSO) $\delta$ 24.3 (Ar–$CH_3$), 117.1 (Ar–C), 122.2 (Ar–C), 128.7 (Ar–C), 137.0 (C–Cl), 153.2 (C of imidazole) 164.6 (HC=N). LC–MS: *m*/*z* 394.40 (M), Anal. calcd. for $C_{21}H_{15}N_5O_3$: C, 75.85; H, 4.58; N, 10.61%. Found: C, 75.87; H, 4.61; N, 10.59%. Scheme 1.

**Acknowledgments**

This research work is financially supported by the Council of Scientific & Industrial Research (CSIR), New Delhi 110 012. (01(2301)/09/EMR-II dated 19-03-2009).

**References**

Alizadeh, M.H., Razavi, H., Bamoharram, F.F., Daneshvar, K., 2003. J. Mol. Catal. 206, 89.

Anastas, P.T., Warner, J.C., 1998. Theory and practice, Green Chemistry. Oxford University Press, Oxford, UK.

Chohan, Z.H., Arif, M., Shafiq, Z., Yaqub, M., Supuran, C.T., 2006. J. Enzyme Inhib. Med. Chem. 21, 95.

Cimerman, Z., Miljanic, S., Galic, N., 2000. Croatica Chemica Acta 73, 81.

Clark, J.H., 2001. Pure Appl. Chem. 73, 103.

Clark, J.H., 2002. Acc. Chem. Res. 35, 791.

Cole, E.R., Crank, G., Salam-Sheikh, A., 1974. J. Agric. Food Chem. 22, 918.

Devassy, B.M., Halligudi, S.B., Elangovan, S.P., Ernst, S., Hartmann, M., Lefebvre, F., 2004. J. Mol. Catal A: Chem. 221, 113.

Gadekara, L.S., Maneb, S.R., Katkara, S.S., Arbada, B.R., Landea, M.K., 2009. Cent. Eur. J. Chem. 7, 550.

Heravi, M.M., Sadjadi, S., Oskooie, H.A., Shoar, R.H., Bamoharram, F.F., 2008. Catal. Commun. 9, 504.

Heravi, M.M., Sadjadi, S., 2009. J. Iran. Chem. Soc. 6, 1.

Hubschwerlen, C., Pfiieger, P., Specklin, J.L., Gubernator, K., Gmunder, H., Angehrn, P., Kompis, I., 1992. J. Med. Chem. 35, 1385.

Iemura, R., Kawashima, T., Fukuda, T., Ito, K., Tsukamoto, G., 1986. J. Med. Chem. 29, 1178.

Jarrahpour, A.A., Jalbout, A.F., Rezaei, S., Trzaskowski, B., 2006. Molbank, 455.

Kruse, L.I., Ladd, D.L., Harrsch, P.B., McCabe, F.L., Mong, S.M., Faucette, L., Johnson, R., 1989. J. Med. Chem. 32, 409.

Kubo, K., Kohara, Y., Yoshimura, Y., Inada, Y., Shibouta, Y., Furukawa, Y., Kato, T., Nishikawa, K., Naka, T., 1993. J. Med. Chem. 36, 2343.

Kuhler, T.C., Swanson, M., Shcherbuchin, V., Larsson, H., Mellgard, B., Sjostrom, J.E., 1988. J. Med. Chem. 42, 1777.

Labanauskas, L.K., Brukstus, A.B., Gaidelis, P.G., Buchinskaite, V.A., Udrenaite, E.B., Dauksas, V.K., 2000. Pharm. Chem. J. 34, 353.

Mederski, W.K.R., Dorsch, D., Anzali, S., Gleitz, J., Cezanne, B., Tsaklakidis, C., 2004. Bioorg. Med. Chem. Lett. 14, 3763.

Pandeya, S.N., Sriram, D., Nath, G., De Clercq, E., 1999. IL Farmaco 54, 624.

Pathak, P., Jolly, V.S., Sharma, K.P., 2000. Oriental J. Chem. 16, 161.

Perry, B.F., Beezer, A.E., Miles, R.J., Smith, B.W., Miller, J., Nascimento, M.G., 1988. Microbois 45, 181.

Porcari, A.R., Devivar, R.V., Kucera, L.S., Drach, J.C., Townsend, L.B., 1998. J. Med. Chem. 41, 1252.

Roth, T., Morningstar, M.L., Boyer, P.L., Hughes, S.H., Buckheit, R.W., Michejda, C.J., 1997. J. Med. Chem. 40, 4199.

Singh, P., Goel, R.L., Singh, B.P., 1975. J. Indian Chem. Soc. 52, 958.

Samadhiya, S., Halve, A., 2001. Orient. J. Chem. 17, 119.

Taggi, A.E., Hafez, A.M., Wack, H., Young, B., Ferraris, D., Lectka, T., 2002a. J. Am. Chem. Soc. 124, 6626.

Taggi, A.E., Hafez, A.M., Wack, H., Young, B., Ferraris, D., Lectka, T., 2002b. J. Am. Chem. Soc. 124, 6635.

Townsend, L.B., WO Patent 1996, 96/07646.

Veerakumari, L., Munuswamy, N., 2000. Vet. Parasitol. 91, 129.